

# Nonequilibrium work equalities in isolated quantum systems

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We briefly introduce the quantum Jarzynski and Bochkov-Kuzovlev equalities in isolated quantum Hamiltonian systems, which includes the origin of the equalities, their derivations using a quantum Feynman-Kac formula, the quantum Crooks equality, the evolution equations governing the characteristic functions of the probability density functions for the quantum work, the recent experimental verifications. Some results are given here first time. We particularly emphasize the formally structural consistence between these quantum equalities and their classical counterparts, which shall be useful in understanding the existing equalities and pursuing new fluctuation relations in other complex quantum systems.

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## I. INTRODUCTION

According to the second law of thermodynamics (the 2nd law) [1], for an arbitrary isothermal process that starts and ends in thermal equilibrium states, the work  $W$  done on the macroscopic system is always larger than or equal to the change of the free energy of the system,  $\Delta G$ , and the equality only holds for a reversible process. With the statistical interpretation of the work, this statement is argued to be still valid even in small systems with significantly fluctuation, *i.e.*, mathematically,

$$\langle W \rangle \geq \Delta G, \quad (1)$$

where  $\langle \rangle$  denotes an average over all nonequilibrium processes of the system undergone during the time interval.

Although the work principle (1) is rigidly established and widely accepted in modern statistical physics and thermodynamics, the 2nd law provides little information about the characteristics of the fluctuation of Eq. (1) in far from equilibrium regime. This situation was not changed until Jarzynski in 1997 found an important equality that is now called Jarzynski equality (JE) [2, 3]:

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta G}, \quad (2)$$

where the nonequilibrium processes or trajectories of an isolated system in the phase space  $\Gamma=(p, q)$  start from the thermal equilibrium state with Hamiltonian  $H(\Gamma, 0)$  at inverse temperature  $\beta$  and then are completely controlled by a time-dependent Hamiltonian  $H(\Gamma, t)$  up to the final time  $t_f$ ,

$$W = \int_0^{t_f} \partial_\tau H(\Gamma(\tau), \tau) d\tau \quad (3)$$

is the work done by external agent on the system,  $\Delta G=G(t_f)-G(0)$ , and  $G(t)$  is the free energy of the system with the quenched Hamiltonian  $H(\Gamma, t)$  at the same inverse temperature. Using the Jensen's inequality, we can reobtain the inequality (1) from the equality (2). Several experiments including single-molecule manipulation techniques have confirmed this equality [4–6]. The intensive interest in the JE also revived another very analogous equality found by Bochkov and Kuzovlev (BKE) [7–10] in the 1970's, when they generalized the fluctuation-dissipation theorems (FDTs) [11–13] into the nonlinear case:

$$\langle e^{-\beta W_0} \rangle = 1. \quad (4)$$

In contrast to the JE, what they were concerned about is a system described by a Hamiltonian  $H_0(\Gamma)$  that is perturbed by a dynamic driven field  $X(t)$ , *i.e.*, the total Hamiltonian is  $H(\Gamma, t)=H_0(\Gamma)-X(t)Q(\Gamma)$ , where  $Q(\Gamma)$  is a conjugate

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generalized coordinate [14], and they defined an alternative work as

$$W_0 = \int_0^{t_f} X(\tau) \dot{Q}(\Gamma(\tau)) d\tau, \quad (5)$$

where the dot denotes the time derivative  $d/d\tau$ . At time 0, the field  $X(0)$  is zero and the system is at the thermal equilibrium with the Hamiltonian  $H_0$  at the inverse temperature  $\beta$ . The BKE (4) immediately leads into an inequality  $\langle W_0 \rangle \geq 0$ , which is nothing just the Kelvin-Planck statement of the 2nd law. The two work equalities are generally not equivalent [15, 16]. Following Jarzynski [15], we call Eqs. (3) and (5) the inclusive work and exclusive work, respectively. These work equalities together with the celebrated fluctuation theorem found earlier by Evans *et al.* [17] in 1993 triggered the enthusiasm of research in the 2nd law in small systems and the fluctuation of nonequilibrium processes, and finally led into a discovery of a series of exact and asymptotic equalities about statistics of various entropy production or dissipated work [2, 3, 7–10, 17–33]. Now these results are all termed “fluctuation relations”. Because the fluctuation relations are related to energy, dissipation, and information during the manipulation and control of small thermodynamic systems, they also attract considerable interdisciplinary interest, *e.g.*, the molecular biophysics and nanoscience [34–36].

With the clarification for the classical systems, in the past decade, extending the classical fluctuation relations into the nonequilibrium quantum regime has been attracting intensive interest and significant progresses have been achieved [7–10, 37–74]. The major challenge of the generalization is that, the key concepts including various classical thermodynamic quantities and classical mechanical picture have to be reconsidered very carefully in the quantum case. For instance, the meanings of the work, heat, and entropy production become ambiguous, whereas the very useful concept of the trajectory in the phase space does not even exist in quantum mechanics. Currently, these research effort is still ongoing. By comparison, for the relatively simple isolated quantum systems, there is a clear consensus about the quantum-extended work equalities [37–39, 47, 48, 53], which is also the object of the current review.

In the literature, there have existed many excellent reviews on the work equalities [34, 62, 75–80]. In particular, Campisil *et al.* [80] systematically reviewed the quantum work equalities just two years ago. Hence, it is very challenging for us to present another one with very fresh perspective for the simple isolated quantum systems. Even though, we do so on the basis of the following several considerations. First, we want to introduce this very active field to the readers who are not experts but are still very interested. The isolated Hamiltonian systems are always the best prime for them. Second, we organize this review by emphasizing the mathematical consistence of the work equalities in classical and quantum isolated systems. In our opinion, it is not only a formal interest, which is a complementary to the review by Campisil *et al.* [80], but also provides a plausible scheme for finding new work equalities in other quantum systems [65, 67, 68]. Finally, the experimental verifications of quantum work equalities have obtained important progresses [81] very recently. We clearly see that this advancement brings the quantum work equalities and quantum measurement and quantum information together and is creating a new research direction [82–85].

The organization of this review is as follows. In Sec. (II), we rederive the classical work equalities from a point of view of irreversibility rather than directly using the definitions of the work. We extend this idea into the isolated quantum systems in Sec. (III). In order to establish an quantum analogy with the classical trajectory-version work equalities, in Sec. (IV) we present a quantum Feynman-Kac formula and obtain new expressions for the existing quantum work equalities. Section (V) is about the quantum Crooks equality. In Sec. (VI), we give a method of computing the characteristic functions of the probability density functions (pdfs) of the quantum work by solving the evolution equations. In Sec. (VII) we review recent experimental progresses in verifying the quantum work equalities. We conclude this review in Sec. (VIII).

## II. CLASSICAL WORK EQUALITIES

Conventional derivations of the classical JE and BKE depend on the definitions of the work [2, 3, 7], namely, defining the work first and approving it satisfying some equality later. This scheme was also used in quantum regimes [37–40, 47, 48, 53, 54]. Although defining work is not a fundamental problem in the classical systems, the situation becomes very subtle in quantum mechanics [40, 54]. It is worthy pointing out that the work equalities including other fluctuation relations essentially arise from the irreversible characteristic of the nonequilibrium processes. Hence, defining the irreversibility rather than pursuing the definition of the work shall be universal either in the classical systems or in the quantum systems. This idea has existed in the literature for long time [23, 24, 29, 31, 32, 75, 86–92]. Here we use the idea to reobtain the equalities (2) and (4).

As mentioned previously, the system is initially at thermal equilibrium with a heat bath with the inverse temperature  $\beta$ , and after time 0, the system is immediately disconnected from the bath (or keeps very weak interaction with the bath) and evolves under the Hamiltonian  $H(\Gamma, t)$  up to the final time  $t_f$ . We call this process the forward process.

We then introduce the backward process with the time-reversed Hamiltonian

$$H_R(\Gamma, s) = H(\tilde{\Gamma}, t_f - s) = H(\Gamma, t_f - s), \quad (6)$$

where  $\tilde{\Gamma} = (-p, q)$  and the new time parameter  $s$  is used to distinguish from the forward time  $t$ . We have assumed that the Hamiltonian is time reversible at arbitrary time point as stated by the second equation of Eq. (6). For simplicity, we do not consider the case of the presence of magnetic field. Very importantly, we specifically assume that the backward process starts from another thermal equilibrium state with the Hamiltonian  $H(\Gamma, t_f)$  at the final time  $t_f$  at the inverse temperature  $\beta$ . Let the density functions of the forward and the backward processes be  $\rho$  and  $\rho_R$ , respectively. Since both the processes follow the Hamiltonian dynamics, according to the Liouville theorem, we have

$$\begin{aligned} \rho_R(\tilde{\Gamma}, t_f) &= \rho_R(\tilde{\Gamma}_{t_f}(\Gamma), 0) = \frac{e^{-\beta H(\Gamma_{t_f}(\Gamma), t_f)}}{Z(t_f)} \\ &= e^{-\beta[H(\Gamma_{t_f}(\Gamma), t_f) - H(\Gamma, 0)]} \rho(\Gamma, 0) \frac{Z(0)}{Z(t_f)}, \end{aligned} \quad (7)$$

where  $\Gamma_{t_f}(\Gamma)$  is the solution of the Hamiltonian's equations at time point  $t_f$  starting from the initial phase point  $\Gamma$ , and  $Z(t) = \int e^{-\beta H(\Gamma, t)} d\Gamma = e^{-\beta G(t)}$  is the instantaneous partition function at time  $t$ . We may clearly see that the whole exponential term except for  $\beta$ , which can be also rewritten as Eq. (3), is the change of the energy of the isolated system along the specific process  $\Gamma_\tau(\Gamma)$ . Hence, it is naturally interpreted as the work done on the *whole* system described by  $H(\Gamma, t)$ , i.e., the inclusive work  $W$  [15]. Integrating the both sides of Eq. (7) with respect to  $\Gamma$ , we reobtain the classical JE (2).

The same procedure is as well available for obtaining the classical BKE (4), where the Hamiltonian is specifically  $H(\Gamma, t) = H_0(\Gamma) - X(t)Q(\Gamma)$ . The only change is that, although the backward process in this case is still under the control of the Hamiltonian defined by Eq. (9), its initial condition is the same with that of the forward process, i.e., the thermal equilibrium state with the Hamiltonian  $H_0(\Gamma)$ . Assume that the density function of the backward process is  $\rho_R^0$ , we have

$$\begin{aligned} \rho_R^0(\tilde{\Gamma}, t_f) &= \rho_R^0(\tilde{\Gamma}_{t_f}(\Gamma), 0) = \frac{e^{-\beta H_0(\Gamma_{t_f}(\Gamma))}}{Z(0)} \\ &= e^{-\beta[H_0(\Gamma_{t_f}(\Gamma)) - H_0(\Gamma, 0)]} \rho(\Gamma, 0). \end{aligned} \quad (8)$$

It is not difficult to see that the exponential term except  $\beta$ , which can be rewritten as Eq. (5), is the change of the energy of the system described by the *bare* Hamiltonian  $H_0(\Gamma)$ . Hence, it was called the exclusive work  $W_0$  [15]. Integrating the both sides of Eq. (8) with respect to  $\Gamma$  leads into the classical BKE (4). The relationship between these two work definitions and work equalities for the classical systems have been discussed in detail [15, 16].

The careful reader may notice that there is no sign of irreversibility in the above discussion. However, the irreversibility indeed comes from the preparation of the thermal equilibrium initial state of the backward process [77]. At the final time  $t_f$ , we need fix the Hamiltonian function at time  $t_f$  and reconnect the system to the heat bath. A relaxation then occurs irreversibly up to the establishment of the thermal equilibrium state. One may prove that  $\langle W \rangle - \Delta G$  is the total entropy production of the whole process including the relaxation [91].

### III. QUANTUM WORK EQUALITIES

The above derivations of classical work equalities can be straightforwardly extended into the isolated quantum systems even if we do not know exactly what a quantum work means. Because we now consider the quantum case, the Hamiltonian functions and the density functions for the classical system are replaced by the Hamiltonian operators  $H$  and density operators  $\rho$ . In order to avoid too many notations, we use the same symbols but without the phase coordinate  $\Gamma$ , which shall not cause confusion in understanding. Analogous to the classical case, we first introduce the backward quantum process of the forward process governed by the Hamiltonian operator  $H(t)$ . Its Hamiltonian is related to the forward one as follows:

$$H_R(s) = \Theta H(t_f - s) \Theta^{-1} = H(t_f - s), \quad (9)$$

where  $\Theta$  is the time-reversal operator. Because the evolutions of both processes are unitary, according to the quantum Liouville theorem, we have

$$\begin{aligned}\rho_R(t_f) &= U_R(t_f)\rho_R(0)U_R^\dagger(t_f) = \Theta U^\dagger(t_f)\Theta^{-1} \frac{e^{-\beta H(t_f)}}{Z(t_f)} \Theta U(t_f)\Theta^{-1} \\ &= \Theta U^\dagger(t_f) e^{-\beta H(t_f)} U(t_f) e^{\beta H(0)} \rho(0) \Theta^{-1} \frac{Z(0)}{Z(t_f)},\end{aligned}\quad (10)$$

where  $U(t)$  and  $U_R(s)$  are the time evolution operators of the forward and backward quantum processes, respectively, which satisfy a general relationship,  $U_R(s) = \Theta U(t_f - s) U^\dagger(t_f) \Theta^{-1}$  [57, 66], *i.e.*, the microreversibility for the isolated quantum system [80]. Making traces of the both sides, we reobtain the quantum Jarzynski equality (QJE) for the isolated system [37–39]:

$$e^{-\beta \Delta G} = \text{Tr}[U^\dagger(t_f) e^{-\beta H(t_f)} U(t_f) e^{\beta H(0)} \rho(0)] = \langle e^{-\beta H^H(t_f)} e^{\beta H(0)} \rangle, \quad (11)$$

where the superscript H denotes the Heisenberg picture. The reasons why it is called an equality about the *quantum* work and why it is physically important shall become explicit after we rewrite the right hand side of the equality in the energy representation [93]:

$$\sum_{m_0, n_{t_f}} p_{\text{eq}}(m_0, 0) |\langle n_{t_f} | U(t_f) | m_0 \rangle|^2 e^{-\beta[\varepsilon_n(t_f) - \varepsilon_m(0)]} = \sum_{m_0, n_{t_f}} p_{\text{eq}}(m_0, 0) p(n_{t_f}, t_f | m_0, 0) e^{-\beta w} = E[e^{-\beta w}], \quad (12)$$

where the Hamiltonian  $H(t)$  is assumed to have discrete transient eigenstates and eigenvalues:  $H(t)|n_t\rangle = \varepsilon_n(t)|n_t\rangle$ . Equation (12) presents a two energy measurement interpretation for Eq. (11). In the first measurement of the system, the outcome of the energy is one of the eigenvalues  $\varepsilon_m(0)$  with the probability  $p_{\text{eq}}(m_0, 0) = e^{-\beta[\varepsilon_m(0) - G(0)]}$ . According to the postulates of quantum mechanics [94], the system is projected into the eigenstate  $|m_0\rangle$ . With time increasing, the system evolves up to the second energy measurement performed at the final time  $t_f$ , which produces an eigenvalue  $\varepsilon_n(t_f)$  with the conditional probability  $p(n_{t_f}, t_f | m_0, 0)$  that is absent in the classical isolate system. Due to energy conservation,  $w$  is very reasonably interpreted as the inclusive work for a specific quantum process. Using the Jensen's inequality, one immediately recovers the work principle  $E[w] \geq \Delta G$ . The definition of the work in quantum case using the two energy measurement scheme was originally proposed independently by Piechocinska [37] and Kurchan [38] in 2000. The idea was further generalized [39] and then was highly developed in a series of articles by Talkner, Hänggi and their coauthors [47–53]. A summary of the later reference may refer to their review article [80]. Even though, our derivation (10) shows that the quantum measurement concept is not always essential for pursuing the work equality.

Using the same procedure, we can reobtain the quantum BKE (QBKE) [53]:

$$\begin{aligned}1 &= \langle e^{-\beta H_0^H(t_f)} e^{\beta H_0} \rangle = \sum_{m, n} p_{\text{eq}}(m, 0) |\langle n | U(t_f) | m \rangle|^2 e^{-\beta[\varepsilon_n - \varepsilon_m]} \\ &= \sum_{m, n} p_{\text{eq}}(m, 0) p(n, t_f | m, 0) e^{-\beta w_0} = E[e^{-\beta w_0}],\end{aligned}\quad (13)$$

where the Hamiltonian  $H_0$  is also assumed to have discrete eigenstate and eigenvalue:  $H_0|n\rangle = \varepsilon_n|n\rangle$ . We see that the two energy measurement scheme is as well available and the work principle  $E[w_0] > 0$  is easily derived.

In the efforts of establishing the quantum work equalities, there were several alternative definitions about the work in quantum regime. Bochkov and Kuzovlev [7, 8] firstly attempted to define a work operator  $\hat{W}_0(t_f) = H_0^H(t_f) - H_0$  in order to extend their equality to quantum case. Yukawa [40], Allahverdyan and Nieuwenhuizen [54] defined another quantum work operator  $\hat{W}(t_f) = H^H(t_f) - H(0)$ . Although the averages of both definitions consist with the average of the work definitions using the two energy measurement scheme, they usually do not lead into correct predictions about the fluctuations of the work and explicit equalities. The fundamental reason is that the work is a quantity about process rather than about the state of the system. Hence, “work is not an observable” [47].

#### IV. QUANTUM FEYNMAN-KAC FORMULA

When we compare the classical and quantum JEs, we notice that there is not a trajectory version for the later. It is not surprising since the classical phase space does not play role in quantum mechanics because of Heisenberg's uncertainty principle [94]. Even though, we are still curious whether there exists an unknown mathematical formula that is equivalent to Eq. (11) but might be more fundamental. Recalling the derivations of the classical JE, a

method on the basis of the celebrated Feynman-Kac formula [95, 96] is very intriguing [3, 15, 87, 89–91, 97, 98]. The formula is valid for the very general Markovian processes including the Hamiltonian mechanics [99]. Importantly, the Feynman-Kac formula is essential the abstract Dyson series [94] represented in the phase or configuration spaces [100]. Hence, we may expect that the method has a quantum-mechanical counterpart, which has been done by Chetrite and Mallick [65], and one of the authors [66], independently. Here we briefly review their results.

Let us consider the following operator equation:

$$\rho_R(s) = \Theta R(t', t_f) \rho_{\text{eq}}(t') \Theta^\dagger, \quad (14)$$

where the parameter  $t' + s = t_f$ , and the instantaneous thermal equilibrium density matrix  $\rho_{\text{eq}}(t') = e^{-\beta H(t')} e^{\beta G(t')}$ . Not strictly speaking, The operator  $R(t', t_f)$  measures the difference between the two density matrixes; see an analogy in the classical system [88]. If  $s = t_f$  Eq. (14) reduces to Eq. (10) due to  $\rho_{\text{eq}}(0) = \rho(0)$ . It is not difficult to prove that,  $R(t', t_f)$  satisfies an evolution equation given by

$$\begin{cases} i\hbar \partial_{t'} R(t', t_f) = [H(t'), R(t', t_f)] - \mathcal{W}_{t'} R(t', t_f) \\ \quad = [H(t'), R(t', t_f)] - i\hbar R(t', t_f) \partial_{t'} \rho_{\text{eq}}(t') \rho_{\text{eq}}^{-1}(t'), \\ R(t_f, t_f) = I, \end{cases} \quad (15)$$

where  $I$  is the identity matrix, and we introduce a superoperator  $\mathcal{W}$ , the action of which on an operator is a multiplication from its right-hand side. Using the Dyson series and time evolution operator  $U(t)$ , we obtain the formal solution for Eq. (15) [65–67]:

$$\begin{aligned} R(t', t_f) &= [G^*(t', t_f) + \sum_{n=1}^{\infty} \int_{t'}^{t_f} dt_1 \cdots \int_{t_{n-1}}^{t_f} dt_n \prod_{i=1}^n G^*(t_{i-1}, t_i) \mathcal{W}_{t_i} G^*(t_n, t_f)] R(t_f, t_f) \\ &= U(t') \mathcal{T}_+ e^{(i\hbar)^{-1} \int_{t'}^{t_f} d\tau U^\dagger(\tau) \partial_\tau \rho_{\text{eq}}(\tau) \rho_{\text{eq}}^{-1}(\tau) U(\tau)} U^\dagger(t'), \end{aligned} \quad (16)$$

where the adjoint propagator  $G^*(t_1, t_2) \mathcal{O} = U(t_1) U^\dagger(t_2) \mathcal{O} U(t_2) U^\dagger(t_1)$  ( $t_1 < t_2$ ), and  $\mathcal{T}_+$  denotes the antichronological time-ordering operator. We called Eq. (16) the quantum Feynman-Kac formula [66]. Substituting the solution into Eq. (14) at  $t' = 0$  and making traces of both sides, we have

$$1 = \text{Tr}[R(0, t_f) \rho_{\text{eq}}(0)] = \langle \mathcal{T}_+ e^{\int_0^{t_f} d\tau U^\dagger(\tau) \partial_\tau \rho_{\text{eq}}(\tau) \rho_{\text{eq}}^{-1}(\tau) U(\tau)} \rangle. \quad (17)$$

Although the equivalence of Eqs. (11) and (17) is undoubted, a more insightful way of seeing it is to do series expansion of  $R(0, t_f)$  except for the term  $e^{-\beta \Delta G}$  therein in terms of the inverse temperature  $\beta$ . Such kind of expansion is not essential to be connected with the high temperature series expansion. For instance, the coefficients of the first two orders are

$$\int_0^{t_f} dt_1 \text{Tr}[\partial_{t_1} H(t_1) G(t_1, 0) \rho_{\text{eq}}(0)] = \int_0^{t_f} dt_1 \langle \partial_{t_1} H^H(t_1) \rangle = E[w], \quad (18)$$

$$\begin{aligned} &\int_0^{t_f} \int_{t_1}^{t_f} dt_1 dt_2 \text{Tr}[\partial_{t_2} H(t_2) G(t_2, t_1) \partial_{t_1} H(t_1) G(t_1, 0) \rho_{\text{eq}}(0)] + \frac{1}{2} \int_0^{t_f} dt_1 \text{Tr}[[H(t_1), \partial_{t_1} H(t_1)] G(t_1, 0) \rho_{\text{eq}}(0)] \\ &= \int_0^{t_f} \int_{t_1}^{t_f} dt_1 dt_2 \langle \partial_{t_2} H^H(t_2) \partial_{t_1} H^H(t_1) \rangle + \frac{1}{2} \int_0^{t_f} dt_1 \langle [H^H(t_1), \partial_{t_1} H^H(t_1)] \rangle = \frac{1}{2} E[w^2], \end{aligned} \quad (19)$$

respectively, where the system's propagator is  $G(t_2, t_1) \mathcal{O} = U(t_2) U^\dagger(t_1) \mathcal{O} U(t_1) U^\dagger(t_2)$ . We see that the double integral of Eq. (19) is about the correlation of the operators at two times. Additionally, this equation clearly explains why defining the work operator  $\hat{W}(t_f) = \int_0^{t_f} \partial_\tau H^H(\tau) d\tau$  alone [40, 54] cannot lead into the correct prediction about the second moment of the quantum work except for the specific canonical initial density matrix. Finally, the expansion does not matter with the initial canonical density matrix. Hence, Eqs. (18) and (19) shall be generally valid, *e.g.*, the microcanonical initial condition [49].

The quantum Feynman-Kac formula (16) is also useful to obtain the new expression of the QBKE (13). In this case, Eq. (14) is replaced by

$$\rho_R^0(s) = \Theta R_0(t', t_f) \rho_{\text{eq}}(0) \Theta^\dagger. \quad (20)$$

The reader is reminded that here  $\rho_{\text{eq}}(0) = e^{-\beta H_0} e^{\beta G(0)}$  specifically. We find that the equation of motion for the new operator  $R_0(t', t_f)$  still has the form as Eq. (15) except that  $\mathcal{W}_{t'}$  is changed into  $\mathcal{W}_{t'}^0 = [\rho_{\text{eq}}(0), H_1(t')] \rho_{\text{eq}}^{-1}(0)$  and  $H_1(t) = X(t)Q$ , where  $Q$  is now an operator [66]. Then we have

$$1 = \text{Tr}[R_0(0, t_f) \rho_{\text{eq}}(0)] = \langle \mathcal{T}_+ e^{-\frac{i}{\hbar} \int_0^{t_f} d\tau U^\dagger(\tau) [\rho_{\text{eq}}(0), H_1(\tau)] \rho_{\text{eq}}^{-1}(0) U(\tau)} \rangle. \quad (21)$$

In addition, very similar results like Eqs. (18) and (19) exist as well, where

$$\partial_\tau H(\tau) \rightarrow \frac{i}{\hbar}[H_1(\tau), H_0], \quad H(\tau) \rightarrow H_0, \quad w \rightarrow w_0. \quad (22)$$

Before closing this section, we want point out several observations. First, if we interpret these operators as ordinary functions,  $-i[\dots]/\hbar$  as the Poisson bracket, and the propagators under the classical meaning, Eqs. (17) and (21) automatically become their classical trajectory-versions. Notice that the time-ordering operator will disappear because of the c-number characteristic of the functions. Second, the equations of motion for  $R(t', t_f)$  and  $R_0(t', t_f)$  inspire us finding the equations governing the evolution of the characteristic functions of the quantum work; see the following discussion. Finally and the most importantly, the definitions of  $R(t', t_f)$  and  $R_0(t', t_f)$ , their equations of motion, and the solutions using the Dyson series do not very depend on whether the system is isolated or not. Hence, the currently developed formal apparatus provides us a plausible path beyond the isolated quantum systems [65, 67, 68].

## V. QUANTUM CROOKS EQUALITY

The QJE (11) and QBKE (13) impose strong restrictions on the pdfs of the quantum work. On the basis of previous argument, for instance, it is easy to see that the pdf of observing a certain value of the inclusive work  $W$  is

$$p(W) = \sum_{m_0, n_{t_f}} p_{\text{eq}}(m_0, 0) p(n_{t_f}, t_f | m_0, 0) \delta(W - w). \quad (23)$$

Using the microreversibility for the isolated quantum system [80], the quantum Crook equality [23, 24] for the inclusive quantum work was obtained [37–39],

$$p(W) = p_R(-W) e^{-\beta(\Delta G - W)}, \quad (24)$$

where  $p_R(W)$  is the pdf of observing a certain value  $W$  for the backward process.

The presence of the delta-function in Eq. (23) is not always convenient in computing the pdf. An elegant way that bypasses it [7–10, 47, 48] is to calculate the characteristic function of the pdf [101],

$$G(u) = \int dW p(W) e^{iuW} = \langle e^{iuH^H(t_f)} e^{-iuH(0)} \rangle, \quad (25)$$

where  $u$  is real number. The Crooks equality (24) indicates a symmetry about the characteristic function:

$$G(u)Z(0) = G_R(-u + i\beta)Z(t_f), \quad (26)$$

where  $G_R$  is the characteristic function for the backward process [38, 48].

Equations. (23)-(26) have their counterparts in the QBKE case [53]. Here we list the results without explanations. Firstly, the pdf of the exclusive work is defined as

$$p_0(W_0) = \sum_{m, n} p_{\text{eq}}(m, 0) p(n, t_f | m, 0) \delta(W_0 - w_0). \quad (27)$$

Its characteristic function  $G_0(u) = \langle e^{iuH_0^H(t_f)} e^{-iuH_0} \rangle$  possesses a symmetry like Eq. (26):  $G_0(u) = G_R^0(-u + i\beta)$ , where  $G_R^0$  is the characteristic function of the exclusive work for the backward process. This symmetry implies  $p_0(W_0) = p_R^0(-W_0) e^{\beta W_0}$ , where  $p_R^0(W_0)$  is the pdf of the exclusive quantum work for the backward process.

The intriguing quantum work equalities and Crooks equality stimulated interest of calculating the pdfs of the quantum work for concrete physical models [102–106]. Because of the involvement of exponential operators and the time-dependent Hamiltonian, however, this job is very challenging and fewer cases can be solved analytically, e.g., the harmonic oscillator with specific time-dependent angular frequency [103] or driven by classical external force [104].

## VI. EVOLUTION EQUATIONS FOR THE CHARACTERISTIC FUNCTIONS

When we compare the right hand side of the characteristic function (25) with that of the QJE (11), their only distinction is that the inverse temperature  $\beta$  in the latter is changed into  $-iu$ . This observation immediately reminds us that  $G(u)$  can be calculated alternatively by firstly solving an operator  $K(t', t_f; u)$  that satisfies

$$\begin{cases} i\hbar \partial_{t'} K(t', t_f; u) = [H(t'), K(t', t_f; u)] - i\hbar K(t', t_f; u) \partial_{t'} e^{iuH(t')} e^{-iuH(t')}, \\ K(t_f, t_f; u) = I, \end{cases} \quad (28)$$



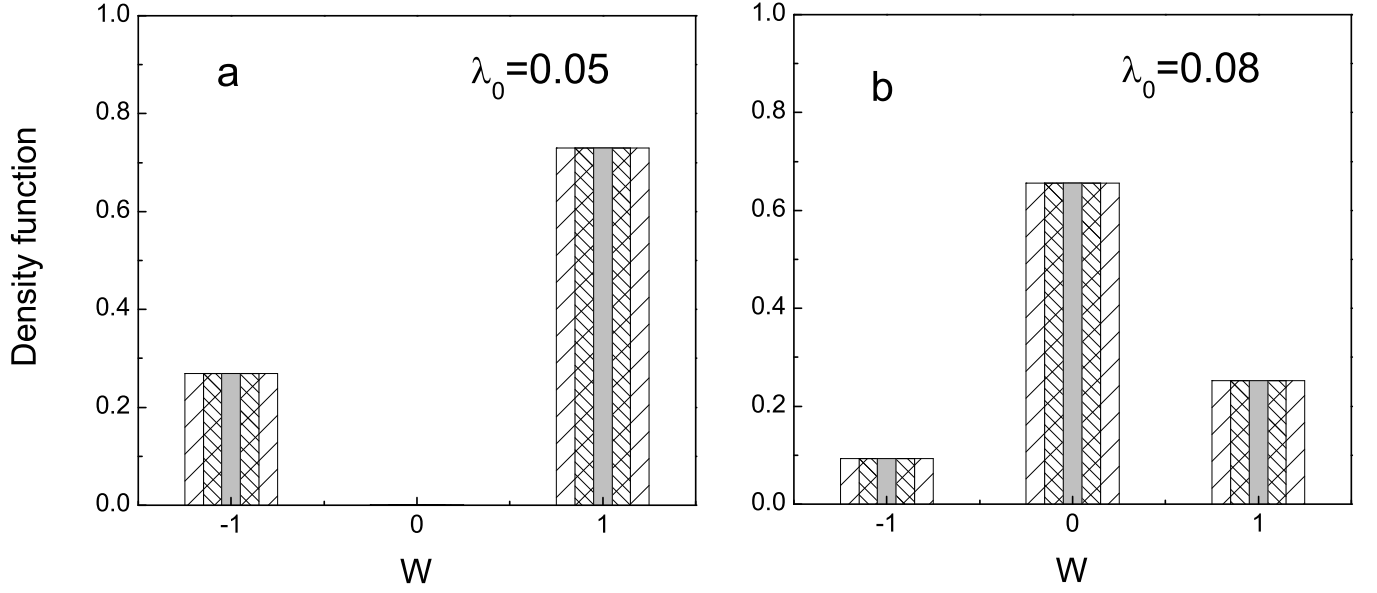


FIG. 1: The pdfs of the quantum work (in unit  $\hbar\omega$ ) for the TLS model (32). The sparse dash bars, solid gray bars, and dense dash bars are calculated by Eqs. (23), (25), and (29), respectively, where  $\beta\hbar\omega=1.0$ ,  $t_f\omega/2\pi=10$ .

and

$$G(u) = \langle K(0, t_f; u) \rangle. \quad (29)$$

This situation is very analogous to the evaluation of an operator's Heisenberg picture: one may either obtain the picture by the definition or solve the Heisenberg's equation [94]. To our knowledge, this result is new in the literature. Similarly, the characteristic function  $G_0(u)$  for the QBKE case also has an alternative expression:

$$G_0(u) = \langle K_0(0, t_f; u) \rangle, \quad (30)$$

where the equation of motion for the operator  $K_0(t', t_f; u)$  is

$$\begin{cases} i\hbar\partial_{t'} K_0(t', t_f; u) = [H(t'), K_0(t', t_f; u)] - K_0(t', t_f; u)[e^{iuH_0}, H_1(t')]e^{-iuH_0}, \\ K_0(t_f, t_f; u) = I. \end{cases} \quad (31)$$

Interestingly, when we recall the characteristic function of the pdf for the classical work, parallel evolution equations have been established early by Imperato and Peliti for the classical stochastic processes [107–109]. Their previous efforts shall be useful in guiding us to further explore the properties of the characteristic functions for the quantum work.

We close this section by numerically solving the pdf of the quantum work for a simple two-level system (TLS) driven by a periodic force, whose Hamiltonian is

$$H(t) = \frac{1}{2}\hbar\omega\sigma_+\sigma_- + \lambda_0 \sin(\omega t)(\sigma_+ + \sigma_-), \quad (32)$$

where  $\sigma_{\pm} = (\sigma_x \pm i\sigma_y)/2$ , and  $\sigma_x$  and  $\sigma_y$  are the Pauli spin matrixes. We specifically choose  $t_f$  to be integer number of cycles so that we do not need distinguish the exclusive or inclusive quantum work. We find the predictions of the three Eqs. (23), (25), and (29) are the same, see Fig. 1.

## VII. MEASUREMENT OF THE QUANTUM WORK

Experimental measurement of the quantum work and verification of the quantum work equalities are very challenging. The crucial requirements include a preparation of an ideally isolated quantum system and two energy measurement at the beginning and the ending of a quantum process. Huber et al. [110] argued that single ion in the linear Paul

trap [111] shall be a remarkable quantum system for testing the QJE. Ion in the Paul trap can be well modelled as a quantum harmonic oscillator [112], and a time-dependent Hamiltonian can be implemented experimentally, *e.g.*, by varying the control voltage of the trap in time [113]. Additionally, there have well-established experimental schemes to create the desired initial thermal equilibrium state, *e.g.*, by performing Doppler cooling [114]. The real experimental obstacle using the trapped ion is how to faithfully read out of the energy eigenstates and their occurring frequencies at the beginning and ending times, respectively. Huber et al. [110] proposed a filtering scheme to implement this task. Its fundamental idea is that the degree of freedom of the oscillator's motion is coupled with the inner electronic state. The latter can be detected by electron shelving method [115]. Even though, such kind of experiments have not been carried out so far.

An alternative very different method was independently proposed by Dorner et al. and Mazzola et al. [82, 83], which is based on Ramsey-like interferometric scheme and bypasses the direct energy projective measurements. Taking the QJE as an example. First, the characteristic function  $G(u)$  (25) is rewritten as

$$G(u) = \text{Tr}[(e^{-iuH(t_f)}U(t_f))^\dagger U(t_f)e^{-iuH(0)}\rho_{\text{eq}}(0)]. \quad (33)$$

Hence, one may regard the terms in the adjoint part to be a time evolution operator of a quantum process at time  $t_f + u$ , the Hamiltonian of which is  $H(t)$  and  $H(t_f)$  before and after time point  $t_f$ , respectively. Similarly, the later part is regarded as the time evolution operator of another quantum process at time  $t_f + u$ . Different from the former, the Hamiltonian is now  $H(0)$  and  $H(t)$  before and after time point  $u$ , respectively. Importantly, Dorner et al. [83] and Mazzola et al. [82] pointed out that these two distinct processes could in fact be merged into one quantum process by coupling the quantum system with the an ancillary qubit  $A$ , whose whole time evolution operator is

$$\bar{U}(u + t_f) = e^{-iuH(t_f)}U(t_f) \otimes |1\rangle\langle 1|_A + U(t_f)e^{-iuH(0)} \otimes |0\rangle\langle 0|_A, \quad (34)$$

where  $\{|1\rangle_A, |0\rangle_A\}$  are the bases of the qubit. By preparing an initial joint density matrix  $\rho_{\text{eq}}(0) \otimes |+\rangle\langle +|_A$ , where  $|+\rangle = (|0\rangle + |1\rangle)\sqrt{2}$ , applying the time evolution operator (34) on the composite system, performing a Hadamard transform at time  $t + t_f$ , one can extract the characteristic function by reading out the state of the ancillary qubit, *i.e.*,

$$\begin{aligned} \rho_A &= \text{Tr}_S[H_A \bar{U}(u + t_f)(\rho_{\text{eq}}(0) \otimes |+\rangle\langle +|_A) \bar{U}^\dagger(u + t_f) H_A] \\ &= \frac{1}{2}(1 + \text{Re}[G(u)]\sigma_z + \text{Im}[G(u)]\sigma_y), \end{aligned} \quad (35)$$

where the trace  $\text{Tr}_S$  is over the concerned quantum system. This scheme is attracting much attention, and particularly, it has been complemented in a spin-1/2 system using a liquid-state NMR device [81]. Finally, we want to mention that, in addition to the above two schemes, their generalizations [84, 85] and other possible experimental schemes [116, 117] were also reported in the literature.

## VIII. CONCLUSION

We close this review by explicitly listing the three key ingredients for establishing the quantum work equalities in the isolated quantum systems. The first ingredient is the microreversibility, which allows us to introduce the time-reversed or the backward quantum processes. We must emphasize that the microreversibility does not always conflict with the irreversible behaviors of nonequilibrium processes at the ensemble level. The isolated systems are very special, since they are reversible at the both levels. The second one is that there exists an instantaneous thermal equilibrium state. In the classical systems, this condition is equivalent to the existence of the instantaneous detailed balance condition. Hence, If the condition is not satisfied, the work equalities may not exist, although one could still define the work. Self-consistently describing the quantum systems by the evolution equations for the systems' density matrix is the last piece of the ingredients. Combining it and the second one, we can derive the equations like Eq. (16) and then the quantum Feynman-Kac formula is obtained. We expect that for some open quantum systems that interact with its environment through energy and matter exchanges, if they possess the these three ingredients, new work equalities could be established accordingly.

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